Comparative leaching characteristics of fly/bottom ashes from municipal solid waste incineration under various environmental stresses

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Abstract

With proper leaching tests, health hazards associated with municipal solid waste incineration (MSWI) ashes, i.e., incineration bottom ashes (IBA) and incineration fly ashes (IFA), can be quantitatively defined. However, it must be coupled with specific environmental scenarios to draw the proper conclusions. Several environmental stresses based on current management of MSWI ashes were herein simulated with laboratory leaching studies to understand their impacts. The impact of bulk metal recovery on the IBA leaching potential was firstly investigated, suggesting the promoted release for certain metals including those with a relative high content (>1000 mg/kg) such as Ba, Cu, Pb and Zn. The impact of seawater was also simulated. Most metal release was altered with the new chemistry established. Batch leaching tests were further performed under both salty and acidic environment to understand their aggregated effects, indicating an overwhelming influence from seawater buffering. Lastly, batch leaching tests of the IBA/IFA mixture were performed under various mass ratios, while data were compared with those by their individuals and the theoretical leaching value, unveiling different leaching characteristics during landfill disposal. Hereby, a comprehensive characteristic metal leaching potential was achieved under various ash managements. It provides insights into environmental risks relevant to their current practices.

Keywords: IBA application; Metal recovery; salty environment; mixed disposal; TCLP

ABBREVIATIONS

MSWI: municipal solid waste incineration

IBA: Incineration Bottom Ashes

IFA: incineration fly ashes

IA: incineration ashes

TCLP: Toxicity Characteristic Leaching Potential

WTE: Waste-to-Energy

DI: deionized

MSW: municipal solid waste

MR: metal recovery

ESP: electrostatic precipitator

APC: air pollution control

LOI: loss on ignition

TC: total carbon

TOC: total organic carbon

ICP-OES: inductively coupled plasma - optical emission spectrometer

ICP-MS: inductively coupled plasma - mass spectrometer

IC: ion chromatography

IBA5: IBA combination sample made of all collection in 5 months

IBA3: IBA combination sample made of all collection in 3 months (the 2^{nd}-4^{th} months)

1. Introduction

MSWI ashes receive various managements in the globe. Several countries have classified the inert wastes which shall go into landfills though, researches have promised their potential reuse via necessary pretreatments (e.g. washing, stabilization and etc.), encompassing substitutes of non-constructive aggregates, sub-base materials for road pavements, embankment fillings, concrete production or marine applications (e.g. coast erosion protection) [1-5]. Concurrently, many landfills keep (or used to) receiving incineration ashes (IA) for mixed dumping (e.g. the Semakau landfill in Singapore) or co-dumping with other inert solid wastes (e.g. the old Lorong Halus dumping ground in Singapore), which may cause secondary pollution for its further redevelopment [6] as a result of their metal leaching [7, 8]. As such, various leaching methods were developed to assess the ash leaching potential, such as Toxicity Characteristic Leaching Potential (TCLP) method, batch leaching methods and column leaching methods [9-11]. However, the real IA leaching behaviors are both scenario-based and site-specific. One-sided conclusion may herein be drawn provided an option of the improper leaching test. Moreover, surrounding geochemistry in the field changes all the time complicating its leaching potential.

The existing management strategies associated with IBA may be categorized into two scenarios, application or disposal. Based on our earlier review work [1], waste to materials has been extensively coupled in Waste-to-Energy (WTE) plants as demonstrated from EU countries. For instance, recycling of IBA is noted with rapid rise in recent years from Netherland (>90%), Demark (90%), Germany (80%), France (70%), Belgium and the UK (21%) [1]. In particular, the construction usage attracts most investigation [12]. Demonstrative experimental projects were performed in which IBA was used as a substitute of aggregates in asphalt mixtures or sub-base material for roads, embankments and in marine applications and concrete substitutes [2, 4, 5, 13]. Note that, however, landfill disposal is still dominant in many countries or regions for the IBA management, due to concerns in association to the application of IBA, including 1) the lack of regulations on IBA utilization, and 2) unsecured geotechnical requirements [1]. Since the exposure pathway of IBA is completely different between the two scenarios which significantly affects its environmental impact, it is necessary to investigate their respective environmental stresses so as to apprehend the IBA leaching potential and enhance its management.

1.1. Application scenario and the associated environmental stresses

Because of the pozzolanic nature and the similar bulk contents to natural aggregates, bottom ash applications can be categorized into two major areas, terrestrial application for construction or land reclamation [1]. As of present, there is no consensus in regulations on which test ought to be implemented for leachability of heavy metals induced by secondary materials. Aiming to discriminate the different application scenarios, appropriate leaching methods have to be carefully chosen to mimic the in- and off-land applications, respectively. In case of off-land applications e.g. land reclamation near seashores, the metal exposure pathways originated from IBA are actually via seawater hydration followed by dilution during construction while porewater transportation through the porous media (such as sediments) afterwards, as opposed to default deionized (DI) water. Metal leaching behaviors from IBA are different among different leaching methods, while statistically correlated to the leachant types, e.g., seawater or DI water [11]. Indeed, it is unveiled based on our previous publication that under percolation conditions seawater tended to enhance most metal leaching as compared to DI water due to its buffering capacity while such effects become more obvious along with increased L/S ratios [14]. Moreover, the IBA application often comes after a metal recovery process to maximize its valorization [15]. Researchers have found that

Fe and Fe oxides play a role in suppressing Pb leaching in the TCLP tests probably ascribed to redox interactions between Pb and Fe and the sorption of soluble Pb onto Fe oxide surface, as well as the leachate pH before and after metal recovery [16]. Two environmental stresses concerning trace metal leaching potentials were herein investigated, associated with various application scenarios based on designed batch leaching tests: the effects of metal recovery and the leachant type (to differentiate between in- and off-land application), to convey the relevant environmental risks during IA application.

1.2. Disposal scenario and the associated environmental stresses

In parallel with the growing attention on ash utilization, there are tremendous amounts of combustion ashes being disposed into landfills every day, due to government legislations, cost efficiency or long-term concerns on health hazards present to various applications [17-20]. One of the most common practices for IBA landfilling may involve the mixing of municipal solid waste (MSW) for co-disposal [19, 21]. Among the dumping activities, mixed ashes disposal combining IBA and incineration fly ashes (IFA) is another choice, such as in Semakau Island – the only off-land landfill in Singapore [22], as it offers easier handling and less cost. Yet, studies up to dates on mixed ashes disposal were rather limited.

In view of aforementioned, a comprehensive study was conducted on characteristic metal leaching potential of IA under various environmental conditions corresponding to existing ash disposals or applications, to understand the full picture of their environmental impacts. The current paper mainly investigated the metal leaching performance of different IAs based on several external stresses including 1) metal recovery; 2) salty and/or acidic environment, and 3) IBA and IFA mixtures. Noteworthy that the leaching methods adopted in the current study were all laboratory-based subjected to limited scales and simplified environmental boundaries as well. In this regard, data may compromise the real scenarios in the field which would be further verified with the large-scale field trials and monitoring in the future.

2. Methodology

2.1. Ash sampling and preparation

IBAs and IFAs used for this study were collected from incineration plant 1 and 2 in Singapore, respectively. Incineration wastes from plant 1 contains more residential source while those from plant 2 contains more industrial source [23]. IBA sampling scheme from plant 1 contained 22 daily samples in the first collection month followed by 14 weekly samples afterward, yielding a total number of 36 samples. IBAs, a mixture of grate sifting, boiler and economizer ash, and grate ash combined at the water quenching tank, were collected directly from ash conveyor of plant 1 before any metal recovery (MR) processes. Approximately 50 kg of freshly quenched IBA was collected for each sample. The IBA was immediately dried at 40 ^{o}C for 3 days upon collection. The weight of the oversized fraction (<1%), i.e. >50 mm, was not used in the study. Ferrous metals were removed using a customized magnetic separator while non-ferrous metals by hand. 36 IBA samples were mixed in equal portions to create the single sample called IBA5 (collection in 5 months) (Table S1). IBA5_MR was then derived after ferrous and non-ferrous metal recovery. IBA3_MR were obtained by mixing equal portions of 3 month IBA samples (weekly sample 1-12 in 2nd-4th collection months) followed by metal recovery process. Before leaching tests were performed, All IBAs (including all subsamples) were crushed with a jaw crusher to below 4 mm (BB 100, Retsch).

Two types of IFAs were collected from incineration plant 2, including one from electrostatic precipitator (ESP) and one from air pollution control (APC). Configuration of the whole incineration process relevant to plant 2 is depicted in Figure S1, whereas lime is employed after ESP for acid gas neutralization followed by precipitation [23]. APC residue is achieved with bag filter with catalyst coating on the surface to remove any dioxin generated from combustion. For preparation of the mixed ashes, ESP and APC residues were firstly mixed at equal portion to form the mixed IFA, then mixed with IBA5_MR at mass ratios of 1:3.5, 1:4 and 1:4.5, respectively, to obtain the designated IBA/IFA mixtures. Normality tests for characteristics of IBA and IFA used in the experiments were performed by using Anderson-Darling statistic (Table S2). Normal distribution analysis was further carried out with 21 elements from all IAs as shown in Table S2. The sample heterogeneity particularly associated with its metal distribution was herein unveiled. Based on Anderson-Darling normality tests, most *p values* of trace metals were <0.005 for IBAs, suggesting a poor normality. By contrast, most metals distribution (15 metals in ESP residue and 19 metals in APC residue out of 21 metals) from fly ashes possessed a *p values* in the range of 0.05-0.87, suggesting a strong normality. As IBA samples used in the study were combined samples (either 3-month or 5-month combination), we hereby assume that all samples in the current study are not statistically significantly different between each other with respects of their element contents and metal leaching potential under identical conditions, allowing to compare their performances excluding their intrinsic heterogeneities.

2.2. Ash characterization

Tests conducted with IBA samples included: moisture content (105 °C for 24 h), loss on ignition (LOI) (550 °C for 2 h), bulk ion leached amounts (Ca, Na, K, Cl, and SO_4), particle size distribution (0-2 mm, 2-4 mm, 4-20 mm, 20-50 mm, and >50 mm), total carbon (TC) and total organic carbon (TOC). TC and TOC were measured with a TOC analyzer (Shimadzu). Rotor mill (ZM 200, Retsch) was used to reduce the IBA sample size below 150 μm before total element measurement. Total element content of each sample was determined by mixing digestion using HCl/HNO3/HF (US EPA Method 3052). The digested solution was first filtered through 0.45 µm nylon membrane followed by dilutions before analysis for cations using inductively coupled plasma - optical emission spectrometer (ICP-OES) (Optima 8300, Perkin Elmer) or inductively coupled plasma - mass spectrometer (ICP-MS) (Nexlon300D, Perkin Elmer) and anions using ion chromatography (IC882 Compact IC plus, Metrohm). Initially, 21 element were monitored for their total content including, Ag, As, B, Ba, Be, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, Sn, Sr, Tl, V and Zn. Based on literature review and their toxicity to human or marine biota, as well as their concentration in IBA leachate [14], 13 element were shortlisted for subsequent studies, i.e., Ag, As, Ba, Cd, Cr, Cu, Hg, Ni, Pb, Sb, Se, Tl, and Zn.

2.3. Ash leaching studies

Different leaching methods adopted in the paper targeted to simulate the IBA leaching potential when it is applied in various field conditions. The pH-static leaching tests intend to establish the complicated leaching profile as a function of pH values, useful to quantify the IBA leachability. On the contrary, batch leaching tests are static leaching method under natural pH which generates chemical data at equilibrium for mechanistic applications. As compared to the mentioned two methods used for the IBA application scenarios, TCLP method is the most popular method designed to mimic field conditions during landfilling [14].

2.3.1. Batch leaching tests

Batch leaching tests (*EN 12457-2*) were adopted for IBA5 and IBA5_MR, while using both seawater and DI water as the leachants. The ash was mixed with the designed leachant at L/S ratio = 10 L/kg on an end-over-end tumbler for 24 h. The eluate was then vacuum-filtered with the 0.45 µm nylon membrane subjected to analysis using ICP-MS (Nexlon 300D, Perkin Elmer) and IC analyzer (IC882 Compact IC Plus, Metrohm) for respective trace elements and salt contents.

2.3.2. pH-static leaching tests

pH-static leaching tests (*CEN/TS 14429*) were conducted on IBA5_MR and IBA3_MR under different pHs, while with application of seawater and DI, respectively. In the test, separate test portions were leached in parallel at a fixed L/S ratio with leachants containing pre-determined amounts of acid or base in order to reach desired stationary pH values at the end of the extraction period. Each leachant was added in three steps in the beginning of the test. At least 8 final pH-values are required, covering a range of pH 2-12. The tests were carried out at a fixed contact time of 48 h, at the end of which equilibrium condition was assumed for most constituents in the IBA. The leachates were filtered through 0.45 µm nylon membrane. The pH was measured by a G20 Compact Titrator (Mettler Toledo, Greifensee, Switzerland). The leachate was acidified to pH 2 and analyzed for traced metals by ICP-OES or ICP-MS.

2.3.3. TCLP leaching tests

IBA5_MR, IFAs, and the IFA/IBA5 mixtures (1:3.5, 1:4, and 1:4.5) were subject to TCLP test (*USEPA SW-846 Method 1311*), simulating the ash disposal scenario. Extraction fluid was firstly prepared followed by leaching tests of different ashes samples. For preparation of the extraction fluid, 11.4 mL acetic acid was mixed with 1988.6 mL DI water to obtain the final volume of 2 L while the pH of fluid was measured at 2.88 ± 0.05. 100 g of the ash sample was introduced into the leaching bottom followed by 2 L of extraction fluid. The slurry was then constantly agitated via an end-over-end rotator at 30 ± 2 rpm for 18 ± 2 h.

Afterward, the leachate was filtered through a 0.45 μm nylon membrane equipped on the glass filter equipment. The pH was measured by a G20 Compact Titrator (Mettler Toledo, Greifensee, Switzerland). The leachate was acidified to pH ≤2 and analyzed for traced metals by ICP-OES or ICP-MS. For comparison, the theoretical leaching data of IBA5/IFA mixtures due to TCLP test was obtained based on calculations through the following equation,

The theoretical leaching value = the measured leaching value of IBA5_MR × the volume fraction (% w/w) of IBA5_MR + the measured leaching value of IFA × the volume fraction (% w/w) of IFA (Eq. 1).

3. Results and Discussion

3.1. Characterization

The particle size distribution based on all IBAs from plant 1 were found similar to those in natural aggregates, composed of sand (31.09±5.02%), gravels (54.92±7.12%) and coarse gravels (13.99±5.41%) (Fig. 1a). As a matter, the grain sizes of IBA particles varies widely from the submicron to a few centimeters or above [24], which is dependent of the MSW feed composition, particle mixing by moving grates and the subsequent treatment of the bottom ashes [25]. LOI was determined at 1.12±0.74%, within typical values of 1-3% [26]. As compared to the general content of unburned organic matter and organic by-products in total approximately 2-5% w/w in concentration [27], the measured value in our study was lower likely due to the adoption of higher combustion temperature [23]. Moisture content was measured at 18.28±1.50% due to water quenching processes [28, 29]. Such a process may bring water content up to 30% [30]. Ferrous metals in IBAs were estimated using a customized magnetic separator, accounting for 9.87±2.61% by weight, and non-ferrous metals by hand, accounting for 1.32±0.61% by weight (Table S1). Both values fell well within the typical value range of their counterparts based on literatures [31].

Trace metal distribution from each IBA, ESP and APC samples was determined as shown in Fig. 1a. Most metal concentrations from all types of ashes presented a limited fluctuation in their distribution (relative standard deviation (RSD) <0.4 for 12 elements, RSD <1.0 for 18 elements), likely ascribed to the stable combustion from the big-scale incinerator (with a capacity of 4000 ton/day) as well as the relative fixed composition of the feeding stock [32]. Among the 21 trace metals (Ag, As, B, Ba, Be, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sb, Se, Sn, Sr, Tl, V, and Zn) under investigation, most metals from ESP residue (except for B, Be, Co and Sr) presented higher concentrations than those in IBA or APC residue, even up to 10-fold more like As, Sb, Zn, etc. Indeed, it is because of the volatility of most metals in high-temperature combustion chambers (e.g. in form of their chloride or oxide compounds) to induce their more enrichment in fly ashes [32, 33]. APC residue contained the lowest levels of all metals (except that Tl and Zn had a higher distribution than those in IBA), likely due to the diluted concentration during the air pollution control process, whereby lime was introduced and precipitated onto the fly ash particles [34]. As a general approach to precipitate acid gases (e.g. SO_2, NOx, HCl, etc.) generated, the application of lime in the dry or semi-dry scrubbing unit could be around 6-20 kg/ton MSW [35] or up to 40% w/w of APC residue [36].

3.2. Ash application

3.2.1. Effects of metal recovery

Effects of metal recovery were studied using the batch leaching method as shown in Fig. 1b. The absence of ferrous and non-ferrous metals favored more release of As, Cd and Pb while suppressed the release of Sb under both DI water and seawater. There were quite some metals presenting an opposite trend between the two eluates, such as Ba, Cr, Cu, Hg and Zn. It is worth noting that, however, the extent of change for Ba under both DI water and seawater, while As, Cu, Ni and Se under DI water was rather limited as compared to the others suggesting an insignificant effect from metal recovery. Enhanced trace metals release could be affected by two possible mechanisms. Firstly, iron oxides and/or aluminum oxides as the respective primary ferrous and non-ferrous metals, carry a high complexation capacity [16, 37]. As a consequence, such effects were revoked after metal recovery process. Secondly, metal recovery actually concentrated heavy metals present in the left IBA (~10% weight loss) (assuming that metal recovery did not concurrently remove any trace metals in the IBA samples), yielding a higher leaching potential likely driven by the enlarged concentration gradient between solid and liquid phases. Yet, contribution from the latter would be minor, given unproportional more metal release presented by Ba (by 59%), Pb (by 547%) and Zn (by 458%) under DI water. For quantitative understanding of the latter effects on the metal leaching potential, comparison of the relative leaching concentration (%) was also performed (Fig. S2), where similar trends of release were well maintained with all metal ions inspected, suggesting a limited influence from the second mechanism.

Fig. 1

3.2.2. Effects of the salty environment

Olsson et al. [38] has reported that the presence of high salinity resulted in increase of certain metal release from the IBA. Mitrano et al. [39] identified a reverse trend that high salt concentrations are often attributed with particle aggregation and sedimentation. However, seawater in the current study illustrated mixed results (Fig. 1b). To understand the effects induced purely by the salt and to exclude the side effects from metal recovery, comparisons were performed under two groups of testing results, 1) IBA5_DI vs. IBA5_SW, and 2) IBA5_DI_MR vs. IBA5_SW_MR, respectively. Cu, Pb and Zn were highly inhibited while Cd, Hg, Sb and Tl were greatly enhanced for both samples (IBA5_SW and IBA5_SW_MR) under seawater. The high pH buffering capacity by seawater was responsible for above differences [40-43]. As a matter, pH values in the leachate based on seawater dropped 2 units to 9.56, resulting in metal redistribution between solid and liquid phases controlled by their respective solubility until new equilibrium was achieved. Cr presented an opposite trend between the two groups, with a decreased concentration for IBA5 however an increased one for IBA5_MR when switching the eluate from DI water to seawater, suggesting a much stronger influence by metal recovery than salt. Other metals like Ag, As, Ba, and Ni maintained limited release despite of the metal recovery process or salinity, suggesting their retarded sensitivity due to their limited amounts and their distribution [44] (Fig. 1b).

Once IBA mixed with the leachant, hydrolysis began while solubility-controlling mineral phases started to dissolve, controlled by surface reactions via diffusion and ion exchange [45]. It resulted in rapid accumulation of various ions in the solution, such as Ca, Na, K, Mg, SO_4, and Cl [46]. Because of the presence of excessive competitive ions from seawater (Na, K, Mg, SO_4, and Cl), however, mineral dissolution from IBA was suppressed. Alongside, co-dissolution of trace metals was inhibited too [34]. Seawater may thus improve the trace metal stability. Under the circumstances of metal recovery, tremendous Fe/Al oxides were removed from the IBA while their associated adsorption capability (for trace metals) no longer existed, facilitating the metal mobility [47, 48]. Metal leaching potential became more complicated with above two principals against each other.

Synergy effects, in a sense, may indicate the dominant factor. Based on analysis of the respective effect from metal recovery and the salt, metal recovery resulted in the same trends of 4 elements including As, Cd, Pb and Sb despite of the presence of salt or not. Contrastively, seawater resulted in the same trends of 7 elements including Cd, Cu, Hg, Pb, Sb, Tl and Zn despite of the presence of metal recovery or not. As such, the salty environment as compared to the metal recovery process, likely dominated the metal leaching potential from the IBA via its strong impacts on the pH value in the system.

3.2.3. Effects based on combined pH and salt

pH-static leaching tests help to quantitatively understand the metal leaching potential as a function of pH values. It generates the typical characteristic leaching curves associated with different metal species in response to the pH variation, to predict their leaching behaviors when various pH conditions are encountered. Seawater as the leachant for pH-static leaching tests may illustrate a more sophisticated situation where both salts and acids are present, which may occur in typical landfills next to the seashore, for instance the Semakau landfill, which is the only landfill receiving all IA generated from Singapore. It is well known that the presence of seawater would buffer the pH values in the solution while the ionic strength may also affect the metal leaching via significant affecting the surface charge of minerals e.g. hydroxides [14, 34]. To this end, pH-static leaching tests based on IBA5_MR were conducted using seawater as the leachant. For comparison, pH-static leaching tests of IBA3_MR were also carried out using DI water as the leachant. Total metal contents within the two tested samples (IBA3_MR vs. IBA5_MR) suggested that the two ashes were comparable on their metal distribution (Fig. S3).

Fig. 2

Seawater indicated a surprisingly strong buffering capacity on the pH values in the final leachate, being well maintained in the range of 8.0-9.5 (Fig. 2). Under these pH values, we noticed that metal leached amounts were well in line with the leaching trends presented by the pH-static leaching tests using DI water based on IBA3_MR. Under the circumstances, the leaching trends of most metals was well controlled at its lower boundaries of leaching availability. As compared to pH, the influences caused by ionic strength is deemed less significant or in other words, ionic strength may pose its influence indirectly via its buffering capacity rather than any specific and or combined ion species.

3.3. Ash disposal

Two scenarios associated with ash disposal were evaluated, encompassing separate disposal and mixed disposal of IBA and IFA.

3.3.1. Effects of separate disposal

TCLP was adopted to simulate ash disposal scenarios, whereby severe conditions (e.g. low pHs) may be derived from landfill acidification [49-51]. Figure 3 illustrates the leaching potential of IBA5_MR and selected sample of ESP and APC residues, respectively. For comparison, the batch leaching results of IBA5_MR was included. TCLP led to more leached amounts of most metals than those from the batch leaching test, due to adoption of acidic leachant (initial pH = 2.88). In addition, higher L/S ratio = 20 in TCLP also favored the overall metal release [52, 53]. As an exception, Cr exhibited excessive release from the batch leaching test, possibly relevant to its metal speciation of Cr (VI), which possess a higher solubility under a higher pH (12.4) in batch leaching tests [1, 54].

Fig. 3

TCLP presented the highest release of Ag, Ba and Pb in APC residue while Cd, Ni and Zn in IBA, albeit their lower contents than ESP residue (Fig. 1a). Again, it was ascribed to pH-controlling metal solubility in the final solution. In addition, solubility-controlling mineral phases and metal speciation were as always contributable for the final metal concentration in the leachate via surface complexation and possibly competitive adsorption [55-57].

3.3.2. Effects of mixing disposal

Mixed ashes composed of IBA5_MR and ESP/APC residue (1:1) were subjected to TCLP tests, while the leaching data were further compared with their theoretical leaching values, showing in Fig. 4. Mixing ratios investigated in the study from 1:3.5 to 1:4.5 (IFA/IBA5), stood for a minor decrease of the IFA fraction from 28.6% down to 22.2% in the mixture. Hereby, the expected differences between the leaching concentrations were insignificant. Cd, Pb and Zn demonstrated an increased release with the increment of the IBA fraction (from 71.4% to 77.8%), likely associated with its more affluence in IBA. With increased IBA fraction, pH value in the leachate slightly decreased from 7.88 to 7.46. As such, pH was thought to be less significant, yet due to its smaller acid neutralization capacity (*ANC*) under the lower pH, most of metals prone to leach more [40].

Fig. 4

Mixed ash disposal likely caused less environmental impact than disposal individually, as the most leachable metals were surprisingly inhibited. For instance, the release of Cr, Ni, Pb and Zn from the ash mixture were inhibited by up to two orders of magnitude. Such stabilization effects would be attributable for the pozzolanic reactions, similar to the concrete formation processes [58]. Postulated mechanisms were shown in Fig. 5, in which IBA acted as the support material while fly ashes as cementitious stabilizer. In the presence of water, the hydration kinetics of fly ash would be much higher (due to smaller particle size) and yield over saturated calcium ions and hydroxides, which could react chemically with other ingredients from the mixtures (mainly the IBA) to form a stabler matrix of solids that may simultaneously fix more metals. These potential pozzolanic reactions are listed as below [59]:

$$Ca(OH)_2 + SiO_2 \rightarrow (CaO)_x \cdot (SiO_2)_y \cdot (H_2O)_z \quad (1)$$

$$Ca(OH)_2 + Al_2O_3 + H_2O \rightarrow (CaO)_x \cdot (Al_2O_3)_y \cdot (H_2O)_z \quad (2)$$

$$Ca(OH)_2 + Al_2O_3 + Fe_2O_3 + H_2O \rightarrow (CaO)_x \cdot (Al_2O_3)_y \cdot (Fe_2O_3)_z \cdot (H_2O)_w \quad (3)$$

Fig. 5

Correlation coefficients between mixed ash leaching and their respective raw resources were calculated based on 13 metals (Table 1), to indicate the contribution of different ashes on the combined leaching results. With an increased mixing ratio of IBA in the ash mixture, there was a higher correlation coefficient, from 0.324 at IFA/IBA5 ratio of 1:3.5, to 0.884 at ratio of 1:4 and then to 0.981 at ratio of 1:4.5. Low correlation coefficient (at 1:3.5) is beyond expectation since the IBA fraction was still dominant (71.4%). From another point of view, there might be a critical percentage of fly ashes, over which the mixed ash leaching characteristics would predominantly depend on the fly ash properties. Based on the study, fly ashes started to play a significant role when its fraction reached to 25.0-28.6% or more. Indeed, due to the much higher metal contents present in fly ashes (e.g. ESP residue) (Fig. 1a) together with its higher metal leaching potential (Fig. 3), its presence in the ash mixture tends to be more critical to the overall mixed ash performance.

Table 1

4. Conclusion

The recovery process of ferrous and non-ferrous metals from IBA may promote the leaching potential of As, Ba, Cd, Ni, Pb and Zn in DI water, as results of reduced surface complexation availability (primarily based on Fe/Al hydroxides) and the increment of apparent concentrations of investigated metals. Contrastively, Ag, Cr, Cu, Hg, Se and Tl showed statistically insignificantly different between concentrations before and after metal recovery in DI water, suggesting their robust inclusion to various minerals. Certain of these metals (e.g. Pb and Zn) are usually highly accumulated from the primary IBA, hereby indicating a raised health hazard. In the presence of seawater, the leaching potential of most metals become dramatically reduced despite of metal recovery or not (except for Cd, Hg and Sb), likely because of its pH buffering capacity. As such, the salty environment may present a stronger stabilization effects on the trace metals from the IBA than that by metal recovery. Indeed, under the same condition of metal recovery the metal leaching potential with seawater was adversely promoted as opposed to those used to be inhibited (by metal recovery) under DI water, including Ba, Cu, Ni, Se and Zn, suggesting the strong effect induced by pH buffering of seawater which may yield a lower pH and thus higher pH-controlling mineral solubilities. pH-static leaching tests with seawater further unveiled that the final pHs in the system were well maintained in 8.0-9.5, echoing the earlier findings. Under IBA disposal scenarios, the mixed ash disposal improved stabilization of certain metals mainly ascribed to the pozzolanic reactions. As a quick conclusion for the application scenario, land reclamation seems a promising alternative as compared to the other terrestrial applications, since seawater stabilized most heavy metals from otherwise leaching by DI water.

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Legends of Tables and Figures

Table 1. Correlation coefficients of heavy metal leaching concentrations (Ag, As, Ba, Cd, Cr, Cu, Hg, Ni, Pb, Sb, Se, Tl, Zn) between each mixed ash (IFA/IBA5 = 1 : 3.5, 1 : 4 and 1 : 4.5, respectively) and each of their primers (IBA5_MR, ESP and APC residues).

Figure 1. a) Total trace element content of ash samples for IBA5, ESP and APC residues, and b) IBA5 metal leaching potential under i) metal recovery with DI water, ii) metal recovery with seawater, iii) DI water without metal recovery, and iv) seawater without metal recovery, respectively.

Figure 2. Comparison of the IBA leaching potential (IBA5_MR vs. IBA3_MR) based on pH-static leaching studies while using DI water (dots) and seawater (circles) as the leachant, respectively.

Figure 3. Comparison of leaching potential of different ash types (IBA5_MR, ESP and APC residues) between different leaching methods (the batch leaching method EN 12457-2 vs. TCLP method).

Figure 4. Comparison of leaching potential caused by individual ashes (IBA5_MR, ESP and APC residues) and their combinations under various mixing ratios (IFA/IBA5 = 1 : 3.5, 1 : 4 and 1 : 4.5, respectively) (Note: "T" refers to theoretical leaching value calculated by Eq. 1).

Figure 5. Demonstrative potential mechanisms for reduced leaching values from mixed ashes (IBA5_MR with ESP and APC residues).